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(54) PROPYLENE RESIN COMPOSITION

(57)Abstract:
PROBLEM TO BE SOLVED: To provide a propylene resin composition having an excellent impact resistance, especially low temperature impact resistance, flexibility, transparency and heat resistance.
SOLUTION: This propylene resin composition comprises polypropylene component and a propylene ethylene copolymer component and (1) the composition comprises 90-50 mol% of propylene monomer unit and 10-50 mol% of ethylene monomer unit, (2) this composition comprises 20-50 wt.% of a component (A) eluting at ≤20° C in a temperature programmed elution fractionation using O-dichlorobenzene as a solvent, 10-70 wt.% of a component (B) eluting at 20 to 85° C, 3-10 wt.% of a component (C) eluting at 85 to 100° C and 10-50 wt.% of a component (D) eluting at ≥100° C, the sum of the components (A) to (D) is 100 wt.% and (3) the component (C) comprises 80 to 20 mol.% of the propylene monomer unit and 20 to 80 mol.% of the ethylene monomer unit.

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CLAIMS

[Claim(s)]
[Claim 1] It consists of a polypropylene component and a copolymer component of a propylene and ethylene. (1) A 90 - 50-mol % and ethylene monomeric unit 10 - 50-mol %, [a propylene monomeric unit] About the leached moiety which came out, exists and was classified by the temperature temperature up elution separation method using a (2) O-dichlorobenzene solvent The amount of 20-degree-C or more less than 85-degree C leached moiety (B component) 20 to 50% of the weight 10 - 70 % of the weight, [the amount of a less than 20-degree C leached moiety (A component)] The amount of 85-degree-C or more less than 100-degree C leached moiety (C component) 3 - 10 % of the weight, The amount of a leached moiety (D component) 100 degrees C or more is 10 - 50 % of the weight. And the polypropylene resin constituent which the sum total of A component, B component, C component, and D component is 100 % of the weight, and is characterized by a propylene monomeric unit being [a 80 - 20 mol % and ethylene monomeric unit] 20 - 80-mol % for (3) C components.

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DETAILED DESCRIPTION

[Detailed Description of the Invention]
[0001]
[Field of the Invention] This invention relates to the outstanding shock resistance and a resin constituent with flexibility, transparency, and thermal resistance good [having especially low-temperature impact resistance].
[0002]
[Description of the Prior Art] Conventionally, generally the approach of adding ethylene-propylene copolymer rubber, ethylene-butene copolymer rubber, a propylene-butene copolymer, a low consistency, straight chain-like low density polyethylene, etc. as a modifier in crystalline polypropylene is learned as an approach of giving shock resistance to polypropylene.
[0003] However, it could not be satisfied with the conventional constituent which blended such a modifier of shock resistance and transparency at coincidence.
[0004] As compared with that from which the polyolefine system elastomer manufactured by the polymerization method was obtained by the above-mentioned blending method on the other hand, transparency is good. Generally by the manufacture approach by this polymerization method, the two-step polymerization method for performing copolymerization of a propylene and ethylene on a second stage story is performed in the polypropylene component in the first stage story. For example, the method of manufacturing thermoplastic elastomer by the polymerization method is indicated by JP,7-118354,A and JP,10-330430,A, and it is indicated that the propylene ethylene copolymer which has the specific presentation acquired as a result shows good flexibility and transparency.
[0005] However, shock resistance and since especially low-temperature impact resistance was low, as for the polypropylene regin obtained by the above-mentioned approach, the further amelioration was desired.
[0006]
[Problem(s) to be Solved by the Invention] Therefore, the purpose of this invention is to have especially low-temperature impact resistance, and for flexibility, transparency, and thermal resistance offer [the outstanding shock resistance and] a good resin constituent.
[0007]
[Means for Solving the Problem] In the polypropylene resin constituent which has specific crystal nature cloth, it has especially cold shock-proof nature, and this invention persons came to complete a header and this invention for outstanding being shock resistance and flexibility, transparency, and thermal resistance being good, as a result of repeating research, in order to solve the above-mentioned problem.
[0008] Namely, this invention consists of a polypropylene component and a copolymer component of a propylene and ethylene. (1) A propylene monomeric unit is 90 - 50-mol %, and an ethylene monomeric unit is 10 - 50-mol %. (2) about the leached moiety classified by the temperature temperature up elution separation method using O-dichlorobenzene solvent The amount of 20-degree-C or more less than 85-degree C leached moiety (B component) 20 to 50% of the weight 10 - 70 % of the weight, [the amount of a less than 20-degree C leached moiety (A component)] The amount of 85-degree-C or more less than 100-degree C leached moiety (C

component) 3 - 10 % of the weight, The amount of a leached moiety (D component) 100 degrees C or more is 10 - 50 % of the weight. And the sum total of A component, B component, C component, and D component is 100 % of the weight, and a propylene monomeric unit is the polypropylene resin constituent characterized by (3) C components being [a 80 - 20 mol % and ethylene monomeric unit] 20 - 80-mol %.

[0009]

[Embodiment of the Invention] A polypropylene resin constituent consists of a polypropylene component and a copolymer component of a propylene and ethylene in this invention.

[0010] In this invention, a propylene monomeric unit is 90-50-mol %, and an ethylene monomeric unit is 10-50-mol %. If a propylene monomeric unit exceeds 90-mol %, flexibility falls, and in being less than [50 mol %], thermal resistance will fall. When flexibility and heat-resistant balance are taken into consideration, a propylene monomeric unit is desirable and a 88-52-mol % and ethylene monomeric unit is 12-48-mol %. Setting to this invention, temperature temperature up elution separation method (TREF) is Journalof. Applied Polymer Science;AppliedPolymer Symposium It is based on 45 and the approach described by the detail 1-24 (1990).

[0011] That is, it is the approach of carrying out elution to order from a component with the low melting point, and isolating an elution polymer component preparatively, by introducing a hot polymer solution into the column which used diatomaceous earth as a bulking agent first, crystallizing a bulking agent front face sequentially from a component with the high melting point, and then raising column temperature gradually by reducing column temperature gradually. Therefore, in this specification, less than 20-degree C leached moieties are all leached moieties in the temperature up process in which it results [from a low temperature side] in 20 degrees C, in temperature temperature up elution separation method.

[0012] In this invention, the amount of the less than 20-degree C leached moiety (it abbreviates to A component hereafter) classified by the above-mentioned temperature temperature up elution separation method is 20 - 50 % of the weight. If shock resistance and flexibility are spoiled and A component exceeds 50 % of the weight at 20 or less % of the weight, thermal resistance will fall. When shock resistance, flexibility, and heat-resistant balance are taken into consideration, the amount of A components is 25 - 45 % of the weight preferably.

[0013] In this invention, the amount of 20-degree-C or more less than 85-degree C leached moiety (it abbreviates to B component hereafter) is 10 - 70 % of the weight. When the amount of B components is less than 10 % of the weight, shock resistance, flexibility, and transparency fall. Moreover, thermal resistance cannot be satisfied if it exceeds 70 % of the weight. In order to have the outstanding shock resistance and flexibility, transparency, and thermal resistance, the amount of B components is 15 - 65 % of the weight preferably.

[0014] In this invention, a propylene monomeric unit is [the 90-50 mol % and ethylene monomeric unit of the low crystal component which consists of an A component and a B component] 10-50-mol %. When flexibility falls when an ethylene monomeric unit is less than [10 mol %], and an ethylene monomeric unit exceeds 50-mol %, thermal resistance falls. if flexibility and heat-resistant balance are taken into consideration -- the ethylene monomeric unit of a low crystal component -- desirable -- 12-48-mol % -- it is 15-45-mol % more preferably. In this invention, the amount of 85-degree-C or more less than 100-degree C leached moiety (it abbreviates to C component hereafter) is 3 - 10 % of the weight. When the amount of C components is less than 3 % of the weight, shock resistance, especially low-temperature impact resistance fall. Moreover, when exceeding 10 % of the weight, transparency falls. When shock resistance and transparency are taken into consideration, the amount of C components is 3 - 9 % of the weight preferably. In this invention, a propylene monomeric unit is [the 80-20 mol % and ethylene monomeric unit of C component] 20-80-mol %. When the propylene monomeric unit of C component is less than [20 mol %], thermal resistance falls, and in exceeding 80-mol %, shock resistance, especially low-temperature impact resistance fall. if thermal resistance and shock-proof balance are taken into consideration -- the ethylene monomeric unit of C component -- desirable -- 20-75-mol % -- it is 25-70-mol % more preferably. The propylene-ethylene random-copolymer component and ethylene monomeric unit beyond 75 mol % are considered that C component consists of the so-called block copolymer

which the propylene-ethylene copolymer of the two above-mentioned component arranged [the propylene monomeric unit] in mixture with the propylene-ethylene random-copolymer component beyond 90 mol %, or a monad chain here. In this invention, it is thought [shock resistance and] that an ethylene monomeric unit depends especially the improvement in low-temperature impact resistance on the propylene-ethylene random-copolymer component beyond 90 mol %.

[0015] In this invention, the amount of a leached moiety (henceforth D component) 100 degrees C or more is 10 - 50 % of the weight. When the amount of D components is less than 10 % of the weight, while the increase of the adhesiveness of the polypropylene resin powder obtained and manufacture become difficult, it is not desirable in order for thermal resistance to fall. Moreover, when exceeding 50 % of the weight, since shock resistance and flexibility fall, it is not desirable. When productivity, thermal resistance, shock resistance, and flexibility are taken into consideration, the amount of D components is less than 30 % of the weight 10 % of the weight or more more preferably ten to 45% of the weight.

[0016] In this invention, a propylene monomeric unit is the fraction which almost accounts for a total rate, and D component consists of a propylene-ethylene random copolymer to which a 100-95-mol % and ethylene monomeric unit changes [a propylene monomeric unit] from 0-5-mol %.

[0017] In this invention, the sum total of the above-mentioned A component, B component, C component, and D component is 100 % of the weight.

[0018] The manufacture approach of the polypropylene resin constituent of this invention is not limited especially as long as the requirements for this invention are satisfied.

[0019] For example, the approach of manufacturing by carrying out the polymerization of the component of the mode which is independent, respectively about the well-known resin which uses the component of - (D) as a principal component, respectively, or is manufactured by blending so that the conditions of this invention may be satisfied for the well-known resin which contains two or more sorts of the above-mentioned component at a rate of arbitration or (A) (A) - (D) which has said property continuously or gradually is mentioned.

[0020] Among those, the approach of acquiring by carrying out a polymerization continuously or gradually improves the dispersibility of each component, and it is the most desirable in order to discover especially cold shock nature enough, the outstanding shock resistance and. Specifically, the method of obtaining the so-called block copolymer which carried out the polymerization of a propylene-ethylene random-copolymer component while the polypropylene component changed the propylene / ethylene presentation continuously or gradually after the polymerization, and the polypropylene component and the copolymerization component of a propylene and ethylene arranged in distribution or a monad chain micro is desirable in order to demonstrate shock-proof improvement, in addition good transparency.

[0021] The following approaches will be mentioned if the typical approach of the approach of manufacturing the polypropylene resin constituent of this invention by the multistage polymerization is illustrated.

[0022] That is, after carrying out the polymerization of the propylene to the bottom of existence of the following catalyst component [I], [RO], and [Ha], it is the approach of performing random copolymerization of a propylene and ethylene succeedingly.

[**] A titanium compound with well-known the titanium compound [RO] organoaluminium compound [Ha] organic silicon compound above-mentioned titanium compound [I] being used for the polymerization of an olefin is used that there is no limit in any way. Especially, when it is used for the polymerization of a propylene, the titanium compound which can obtain the polymer of high stereoregularity by high yield is desirable. These titanium compounds are divided roughly into a support mold titanium compound and a titanium-trichloride compound. The process of a support mold titanium compound is adopted that a well-known approach does not have a limit in any way. For example, JP,56-155206,A, said -- 56-136806 -- said -- 57-34103 -- said -- 58-8706 -- said -- 58-83006 -- said -- 58-138708 -- said -- 58-183709 -- said -- 59-206408 -- said -- 59-219311 -- said -- 60-81208 -- said -- 60-81209 -- said -- 60-186508 -- said -- 60-192708 -- said -- 61-211309 -- said -- 61-271304 -- said -- 62-15209 -- said -- 62-

11706 -- said -- the approach shown in 62-72702 and this 62-104810 grade is adopted. Specifically, the approach of co-grinding halogenation titanium and a magnesium compound or the method of contacting halogenation titanium, a magnesium compound, and an electron donor in a solvent is listed to the bottom of existence of electron donors, such as the approach of co-grinding a titanium tetrachloride with a magnesium compound like a magnesium chloride, alcohol, the ether, ester, a ketone, or an aldehyde.

[0023] Moreover, alpha, beta, gamma, or delta-3 titanium chloride well-known as a titanium-trichloride compound is mentioned. the preparation approach of these titanium-trichloride compounds -- for example, JP,47-34478,A -- said -- 50-126590 -- said -- 50-114394 -- said -- 50-93888 -- said -- 50-123091 -- said -- 50-74594 -- said -- 50-104191 -- said -- 50-98489 -- said -- 51-136625 -- said -- the approach shown in 52-30888 and this 52-35283 grade is adopted. In order especially to obtain the propylene-ethylene copolymer of this invention also in this, a titanium-trichloride compound is desirable.

[0024] Next, a compound with well-known an organoaluminium compound [RO] being used for the polymerization of an olefin is adopted that there is no limit in any way. For example, trimethylaluminum, triethylaluminum, tree n propyl aluminum, Tree n butyl aluminum, tree i butyl aluminum, tree n hexyl aluminum, Trialkylaluminiums, such as tree n octyl aluminum and tree n DESHIRU aluminum; Diethyl aluminum mono-chloride, Diethyl aluminum mono-halide, such as a diethyl aluminum star's picture; alkylaluminum halide, such as methyl aluminiumsesquichloride, ethylaluminiumsesquichloride, and diethyl aluminum chloride, is mentioned. Alkoxy aluminum, such as mono-ethoxy diethyl aluminum and diethoxy monoethyl aluminum, can be used for others. Also in this, trialkylaluminiums especially trimethylaluminum, triethylaluminum, and tree i butyl aluminum are desirable.

[0025] furthermore, **, such as a cyclic hydrocarbon which an organic silicon compound [Ha] is a chain-like hydrocarbon whose atom linking directly to a silicon atom is the 3rd class carbon although a compound with well-known being used for stereoregularity amelioration of an olefin is adopted that there is no limit in any way, or is the 2nd class carbon, -- in order that the organic silicon compound which has a high substituent may make the stereoregularity of the polypropylene component obtained higher and may discover good thermal resistance, it is desirable. Specifically JI t-butyl dimethoxysilane, t-butyl ethyl dimethoxysilane, JI t-amyl dimethoxysilane, dicyclopenthyl dimetoxysilane, Dicyclohexyl dimetoxysilane, t-butyl methyl dimethoxysilane, t-butyl ethyl dimethoxysilane, cyclopentyl methyl dimethoxysilane, Organic silicon compounds, such as cyclopentyl ethyl dimethoxysilane, cyclopentyl isobutyl dimethoxysilane, cyclohexyl methyldimetoxysilane, cyclohexyl ethyl dimethoxysilane, and cyclohexyl isobutyl dimethoxysilane, can be mentioned. Dicyclopenthyl dimetoxysilane and especially t-butyl ethyl dimethoxysilane are desirable especially. Moreover, these organic silicon compounds can also use two or more sorts for coincidence.

[0026] the combination of the titanium compound [I] used for this invention, an organoaluminium compound [RO], and an organic silicon compound [Ha] -- (1) titanium-trichloride compound-trialkylaluminium -- a kind-organic silicon compound (2) support mold titanium compound-trialkylaluminium-organic silicon compound (3) -- the combination of a **** type titanium compound-3 titanium-chloride compound-trialkylaluminium-organic silicon compound is desirable especially in order to satisfy the requirements for a configuration of the polypropylene resin constituent of this invention. Especially the combination of (1) is suitable in order to obtain the polypropylene resin constituent with which are satisfied of this invention.

[0027] Since the smeariness at the time of setting to this invention, and performing precuring by mixing with propylene independence, ethylene independence or a propylene, and ethylene to the bottom of existence of the above-mentioned catalyst component [I], [RO], and [Ha] reducing the amount of generation of the low-molecular-weight object of the polypropylene resin constituent obtained in advance of this polymerization under existence of each above-mentioned component, and considering as mold goods can be suppressed, it is suitable. The iodine compound which is furthermore shown by the following general formula if needed in addition to the above [I], [RO], and [Ha] [NI]

Since performing precuring by mixing with propylene independence, ethylene independence or a

propylene, and ethylene to the bottom of existence of R-I (however, R being the alkyl group or phenyl group of an iodine atom or carbon numbers 1-7.) can suppress further the smeariness at the time of controlling further generation of the above-mentioned low-molecular-weight object, and using the polypropylene resin constituent obtained as a Plastic solid, it serves as a desirable mode.

[0028] Since the amounts of the above [I] used by precuring of this invention, [RO] and [Ha], and each catalyst component of [NI] used further if needed differ according to the class of catalyst component, and the conditions of a polymerization, they should just determine the optimal amount used beforehand according to these monograph affairs. It will be as follows if the range used suitably is illustrated.

[0029] the operating rate of the organoaluminium compound [RO] used for precuring -- a titanium compound [I] -- receiving -- aluminum/Ti (mole ratio) -- 0.1-100 -- desirable -- the range of 0.1-20 -- the operating rate of an organic silicon compound [Ha] -- a titanium compound [I] -- receiving -- [Ha]/Ti (mole ratio) -- 0.01-100 -- the range of 0.01-10 is preferably suitable respectively. moreover, the operating rate of the iodine compound [NI] used if needed -- a titanium compound [I] -- receiving -- I/Ti (mole ratio) -- 0.1-100 -- the range of 0.5-50 is preferably suitable.

[0030] It is as follows when the iodine compound which can be suitably used by precuring of this invention is shown concretely. For example, they are iodine, a methyl iodide, an ethyl iodide, propyl iodide, iodation butyl, an iodobenzene, p-iodation toluene, etc. Especially a methyl iodide and an ethyl iodide are suitable.

[0031] although the amount of precuring which carries out a polymerization by mixing of propylene independence, ethylene independence or a propylene, and ethylene to the bottom of existence of said catalyst component changes with precuring conditions -- general -- a 0.1 - 250 g/g-Ti compound -- if it is the range of a 1 - 100 g/g-Ti compound preferably, it will come out enough.

[0032] It is possible in the case of precuring to also make hydrogen live together. As for this precuring, it is desirable to make a slurry polymerization usually apply, and as a solvent, it is independent about saturated aliphatic hydrocarbon or aromatic hydrocarbon, such as a hexane, a heptane, a cyclohexane, benzene, and toluene, or can use these mixed solvents.

[0033] This especially precuring temperature has the desirable range of 0-60 degrees C -20-100 degrees C. What is necessary is just to decide on precuring time amount suitably according to the lap in precuring temperature and precuring. Although the pressure in precuring is not limited, in the case of a slurry polymerization, generally, it is about atmospheric pressure -5 kg/cm2G. This precuring may be performed by the batch, the half-batch, and which approach of continuation.

[0034] Subsequently to said precuring, this polymerization is carried out. Under existence of the catalyst content precuring object with which this polymerization was obtained by said precuring, the polymerization of a polypropylene component is performed first and then random copolymerization of propylene-ethylene is carried out. Moreover, although each catalyst component can also use what was added at the time of precuring in the condition as it is, it is desirable except a titanium compound to newly add and to adjust at the time of this polymerization.

[0035] Since the amount and polymerization conditions of each catalyst component of the above [I] used by this polymerization of this invention, [RO], and [Ha] differ from each other according to the class of catalyst component, they should just determine the optimal amount used and polymerization conditions beforehand according to the class of these catalyst components. It will be as follows if the amount and polymerization conditions of a catalyst component which are used suitably are illustrated.

[0036] The above-mentioned thing can use the organoaluminium compound [RO] used by this polymerization that there is no limit in any way. the amount of the organoaluminium compound used -- catalyst content precuring -- a titanium atom in the living body -- receiving -- aluminum/Ti (mole ratio) -- it is -- 1-1000 -- it is 2-500 preferably. Moreover, although this lap changes with these polymerization conditions, generally it is the range of 1000-50000g/g and a

titanium compound.
[0037] The organic silicon compound [Ha] used by this polymerization is adopted that a compound as stated above does not have a limit in any way. the amount of the organic silicon compound used used by this polymerization -- catalyst content precuring -- a titanium atom in the living body -- receiving -- Si/Ti (mole ratio) -- 0.001-1000 -- it is 0.1-500 preferably.
[0038] As for this above-mentioned polymerization, the polymerization of a polypropylene component is carried out first. The polymerization of a polypropylene component supplies the propylene in within the limits and the mixture of ethylene with which are satisfied of the requirements for a propylene independent or this invention, and should just carry them out. If the typical conditions of a polypropylene component polymerization are illustrated, it is suitable for polymerization temperature to adopt from the range of 80 degrees C or less and further 20-70 degrees C. Moreover, hydrogen can also be made to live together as a molecular weight modifier if needed. Furthermore, which approaches, such as a slurry polymerization which uses the propylene itself as a solvent, a vapor phase polymerization, and solution polymerization, may be used for a polymerization again. When the shape of the simple nature of a process, a reaction rate, and corpuscular character of a copolymer to generate is taken into consideration, the slurry polymerization which uses the propylene itself as a solvent is a desirable mode. Which approach of a batch process, a half-batch process, and continuous system is sufficient as a polymerization format. Furthermore, a polymerization can be divided into two or more steps where conditions, such as hydrogen concentration and polymerization temperature, differ, and can also be performed.
[0039] Next, random copolymerization of a propylene and ethylene is performed. Which approach of a batch process, a half-batch process, and continuous system may be used for the random copolymerization of a propylene and ethylene, and it can divide a polymerization into a multistage story and can also carry it out. Moreover, which approach of a slurry polymerization, a vapor phase polymerization, and solution polymerization may be used for the polymerization of this process. It is continuing at the polymerization of said polypropylene component in the case of the slurry polymerization which uses the propylene itself as a solvent, and supplying ethylene gas, and, in the case of a vapor phase polymerization, carries out by supplying the mixed gas of a propylene and ethylene.
[0040] In order to satisfy the weight rate of (A) - (D) component of this invention especially, it is desirable to perform random copolymerization of a propylene and ethylene by the slurry polymerization following the polymerization of a polypropylene component. In that case, ethylene's supplying ethylene improves the dispersibility of each component so that ethylene / propylene presentation ratio may go up continuously, and supply, or ethylene / propylene presentation ratio may change a constant rate gradually continuously, and it is desirable in order to fully demonstrate especially the improvement in a cold shock, in addition good transparency, shock resistance and.
[0041] 80 degrees C or less of polymerization temperature of the random copolymerization of a propylene and ethylene are preferably adopted from the range of 20-70 degrees C. Moreover, hydrogen can also be used as a molecular weight modifier if needed, and the hydrogen concentration in that case can be changed to a multistage story, and can also carry out a polymerization.
[0042] After termination of this polymerization, a monomer can be separated from a polymerization system and the polypropylene resin constituent of this invention can be obtained. This polypropylene resin constituent can perform well-known washing or counterflow washing with a with a carbon number of seven or less hydrocarbon.
[0043] In this invention, although especially the melt flow rate of a polypropylene resin constituent is not limited, generally it is desirable that they are 0.1-100g / 10min. When the above-mentioned melt flow rates are 0.1g / less than 10 min, a moldability falls, when exceeding 100g / 10min, a low-molecular-weight object increases and the fall of physical properties may be caused. if a moldability and physical properties are taken into consideration -- a melt flow rate -- desirable -- 0.1-70g/-- they are 0.1-50g / 10min still more preferably 10 min.
[0044] The melt flow rate can be adjusted and the polypropylene resin constituent of this

invention can also be used so that it may become the melt flow rate demanded by the application etc. if needed. The method of adding organic peroxide to the polypropylene resin powder which might make a small amount of hydrogen live together as an approach of adjusting a melt flow rate, at the time of a polymerization, and adjusting molecular weight by carrying out melting kneading is desirable in respect of reduction of a low-molecular-weight object, and the improvement in a moldability.

[0045] After adding fillers, such as commercial additives, such as an antioxidant, a coloring agent, a thermostabilizer, a chlorine supplement agent, lubricant, an ultraviolet ray absorbent, an illuminant-proof, an antistatic agent, an antifogger, an anti blocking agent, a flame retarder, and a crystalline-nucleus agent, and talc, a magnesium hydroxide, a calcium carbonate, glass, a mica, wood flour, and mixing, you may use by making the polypropylene resin constituent of this invention into a pellet with an extruder.

[0046] Furthermore, to the polypropylene resin constituent of this invention, other resin can be added in the range which does not affect the property of this invention remarkably. For example, alpha olefins more than 90% mol of a propylene, and other than a propylene, For example, 1-butene, 1-pentene, 1-hexene, 1-heptene, The random copolymer not more than [one or more sorts of /, such as 1-methyl-1-pentene,] 10 mol %, High density polyethylene (HDPE), low density polyethylene (LDPE), the line which becomes by copolymerization of ethylene, and C4-C10 -- polyethylene (LLDPE) -- Polyethylene system resin, such as ethylene vinyl acetate copolymerization (EVA) and ethylene methacrylate (EMMA), Ethylene propylene rubber (EPR, EPDM), an ethylene butene-1 copolymer (EBM), Well-known things, such as olefin system elasticity resin, a styrene butadiene block copolymer (SBR), petroleum resin and terpene resin, or those hydrogenation objects, such as a propylene butene-1 copolymer (PBM), can use it without a limit.

[0047] As mentioned above, of course, it can also be used as a modifier of various polyolefines besides other polypropylene regins using properties, such as the property, for example, flexibility, and transparency, that the polypropylene resin constituent of this invention can make this a principal component, and it can be used for a shaping raw material.

[0048] The resin constituent of the polypropylene resin constituent of this invention and other resin is obtained by carrying out melting kneading. Although especially the approach of melting kneading is not limited, it is good to, perform 160-300 degrees C to the bottom of the temperature of 180-270 degrees C preferably for example, using a screw extruder, a Banbury mixer, a roll mill, etc. Moreover, this melting kneading can also be performed under inert gas air currents, such as nitrogen gas. In addition, well-known mixed equipment, for example, a tumbler, a Henschel mixer, etc. can use it before melting kneading that there is no limit in any way.

[0049] Furthermore, it is also possible by mixing it, after each component of the above-mentioned resin constituent etc. blends each component if needed, supplying to a direct making machine and fabricating to acquire a Plastic solid. Moreover, when fabricating a film sheet etc., it can fabricate by the well-known shaping approach. For example, the shaping approaches, such as T-die shaping, calender shaping, and inflation molding, can be used.

[0050] It can also be used by the monolayer besides using it as a film sheet, the polypropylene resin constituent of this invention being able to carry out the laminating of the resin layers, such as ethylene system resin, such as propylene system resin, high density polyethylene, low density polyethylene, straight chain-like low density polyethylene, an ethylene-vinylacetate copolymer, and an ethylene-methacrylic-acid copolymer, and multilayering it.

[0051]

[Effect of the Invention] Further, it excels in flexibility, thermal resistance, and transparency, and moreover, fabrication nature is good and the polypropylene resin constituent of this invention can be used suitable for shock resistance especially low-temperature impact resistance, and the various fields for which conventional thermoplastic elastomer is used. For example, in the injection-molding field, each shaping component, such as wheels, such as various packing and a cleaner, a bumper, and a hose, the various shaping components of OA equipment and skiing shoes, a grip, and roller skating are mentioned in the bumper in autoparts, a mud guard, horn putt, lamp packing, and the household-electric-appliances field. On the other hand as a film

application, a wrap film, a shrink film, A SUTOTCHIRE film, the film for sealants, a sizing film, Adhesive tape, a masking film, the film for agriculture, a medical-application film pasting base material, As a sheet, the film for adhesive bandages, a surface-protection film, a makeup film, etc. A stationery sheet, An occlusion sheet, a desk mat, the sheet for agriculture, a tarpaulin, the interior epidermis material in autoparts, Corrugated one, a window mall, an air duct hose, a weather toss trip, wire covering material, etc. can be used as a building-materials-related sheet suitable [wallpaper, flooring etc.] for a vanity case, a makeup bag, a shipping box, a package bag, a food container, miscellaneous-goods components, a toy, tatami facing, etc. as a Plastic solid.
[0052]
[Example] Hereafter, although an example and the example of a comparison are given and this invention is explained, this invention is not limited to these examples. The measuring method used in the following examples is explained.
[0053] 1) the measurement JEOL [Co., Ltd.] Co., Ltd. make of a propylene monomeric unit and an ethylene monomeric unit -- it computed from 13 C-NMR spectrum measured using GSX-270.

[0054] 2) The judgment approach of A component, B component, C component, and D component (the temperature temperature up elution judgment approach)
The following Measuring conditions performed using MPC[by the YUNIFU Rose company]-1100 mold.
[0055]
Solvent 30 - 60mesh column cooling rate; 5 degrees C/Hr The;O-dichlorobenzene rate of flow ; 1.0 ml/min programming rate ; 40 degrees C / Hr sample concentration ; 0.5wt% sample injection rate; 500microl detector ; An infrared detector, wavelength column of 3.14 micrometers ; phi0.9cmx17.5cm bulking agent ; chromosrb [0056] P 3) Measurement ASTM of a melt flow rate (MFR) It was based on D-1238.
[0057] 4) Shock-proof (Izod impact resistance value) measurement JIS Based on K7110, it measured with the notch in the temperature of -40 degrees C.
[0058] 5) Measurement JIS of flexibility (bending elastic modulus) It was based on K7203.
[0059] 6) Measurement JIS of transparency (Hayes value) It was based on K6714.
[0060] 7) Heat-resistant (BIGATTO softening temperature) measurement JIS Based on K7206, it measured on the conditions of 250g of loads.
[0061] The example of the polypropylene resin constituent of this invention is explained below.
[0062] After nitrogen gas fully permuted the glass autoclave reactor of 1l. of content volume equipped with example 1 (precuring) agitator, hexane 400ml was inserted in. After keeping whenever [reactor internal temperature] at 20 degrees C and adding dicyclopenthyl dimetoxysilane 4.2mmol, ethyl-iodide 21.5mmol, triethylaluminum 21.5mmol, and titanium-trichloride (Marubeni Solvay chemistry company make) 21.5mmol, the propylene was continuously introduced into the reactor for 30 minutes so that it might be set to 3g per 1g of titanium trichlorides. In addition, temperature in the meantime was held at 20 degrees C. After suspending supply of a propylene, nitrogen gas fully permuted the inside of a reactor, and the obtained titanium content polypropylene was washed 4 times by the purification hexane. The polymerization of the 2.7g [per 1g of titanium trichlorides] propylene was carried out as a result of analysis.
[0063] (This polymerization) In addition, the temperature up of the internal temperature of an autoclave was carried out to 55 degrees C so that the concentration in a gaseous phase might become the 2l. autoclave which performed N2 permutation about a liquid propylene and might become 1.0-mol% about 1l., triethylaluminum 0.5mmol, dicyclopenthyl dimetoxysilane 0.25mmol, and hydrogen. Next, after supplying ethylene so that the concentration in a gaseous phase may become 1.5-mol%, in addition, copolymerization of propylene-ethylene was performed for 20 minutes at 55 degrees C 0.06 mmols by making into a titanium trichloride the titanium content polypropylene obtained by precuring (process 1). Subsequently, ethylene was supplied by part for 0.8l./by reference condition, and the polymerization for 120 minutes was performed (process 2). The rate of ethylene gas to the propylene gas in a gaseous phase increased as propylene-ethylene copolymerization time amount made it the increment. In addition, the ethylene gas

presentation in the gaseous phase at the time of reaction termination was 15-mol%. After reaction termination, the unreacted monomer was purged and the polymer was obtained. The obtained polymer was dried at 70 degrees C for 1 hour. The melt flow rates of the obtained polymer were 0.10g / 10min.

[0064] (Molecular weight accommodation) As the 0.2 weight section and organic peroxide, the anti-oxidant was extruded at 230 degrees C using Brabender to it, after carrying out 0.05 weight section addition of the 1 and 3-screw-(t-butyl PAOKI seesaw propyl) benzene and mixing to the obtained polymer, and the pellet was obtained. MFRs were 2.0g / 10 minutes.

[0065] (Component analysis) Measurement of the propylene monomeric unit of the low crystal component which consists of measurement of the propylene monomeric unit of a polypropylene resin constituent and an ethylene monomeric unit, A-D each amount measurement of components, an A component, and a B component, and D component, and an ethylene monomeric unit was performed using the pellet which may have had molecular weight accommodation performed. A result is shown in Table 1.

[0066] (Physical-properties measurement) The test piece for Hayes with a thickness of 1mm was fabricated with 15t injection molding machine using the pellet which may have had molecular-weight accommodation performed with the cylinder temperature of 230 degrees C, and the die temperature of 40 degrees C in a bending modulus of elasticity, BIGATTO softening temperature and the shock-proof test piece, and the list. The measurement result of physical properties is shown in Table 2.

[0067] In molecular-weight adjustment of example 2 example 1, the same actuation as an example 1 was performed except having considered the ethylene amount of supply of this polymerization process 2 as a part for 0.6l./. In addition, the melt flow rates of a 12-mol % and the obtained polymer of the ethylene gas presentation in the gaseous phase at the time of reaction termination were 0.12g / 10min. A result is shown in Tables 1 and 2.

[0068] In example 3 example 1, the same actuation as an example 1 was performed except having considered the ethylene amount of supply of this polymerization process 2 as a part for 1.0l./. In addition, the melt flow rates of a 18-mol % and the obtained polymer of the ethylene gas presentation in the gaseous phase at the time of reaction termination were 0.08g / 10min. A result is shown in Tables 1 and 2.

[0069] After nitrogen gas fully permuted the glass autoclave reactor of 1l. of content volume equipped with example of comparison 1 (precuring) agitator, hexane 400ml was inserted in. After keeping whenever [reactor internal temperature] at 20 degrees C and adding diethyl aluminum chloride 18.5mmol, ethyl-iodide 22.7mmol, diethylene-glycol wood ether 0.18mmol, and titanium-trichloride (Marubeni Solvay chemistry company make) 22.7mmol, the propylene was continuously introduced into the reactor for 30 minutes so that it might be set to 3g per 1g of titanium trichlorides. In addition, temperature in the meantime was held at 20 degrees C. After suspending supply of a propylene, nitrogen gas fully permuted the inside of a reactor, and the obtained titanium content polypropylene was washed 4 times by the purification hexane. The polymerization of the 2.9g [per 1g of titanium trichlorides] propylene was carried out as a result of analysis.

[0070] (This polymerization) In addition, the temperature up of the internal temperature of an autoclave was carried out to 55 degrees C so that the concentration in a gaseous phase might become the 2l. autoclave which performed N2 permutation about a liquid propylene and might become 1.0-mol% about 1l., diethyl aluminum chloride 0.7mmol, butyl-acetate 0.07mmol, dicyclopenthyl dimetoxysilane 0.07mmol, and hydrogen. Next, after supplying ethylene so that the concentration in a gaseous phase may become 1.0-mol%, in addition, copolymerization of propylene-ethylene was performed for 30 minutes at 55 degrees C 0.087 mmols by making into a titanium trichloride the titanium content polypropylene obtained by precuring (process 1). Subsequently, ethylene was supplied by part for 0.5l./by reference condition, and the polymerization for 120 minutes was performed (process 2). The rate of ethylene gas to the propylene gas in a gaseous phase increased as propylene-ethylene copolymerization time amount made it the increment. In addition, the ethylene gas presentation in the gaseous phase at the time of reaction termination was ten-mol%. After reaction termination, the unreacted

monomer was purged and the polymer was obtained. The obtained polymer was dried at 70 degrees C for 1 hour. The melt flow rates of the obtained polymer were 0.03g / 10min.

[0071] (Molecular-weight accommodation) The same actuation as an example 1 was performed to the obtained polymer except having carried out 0.08 weight section addition of the anti-oxidant 0.2 weight section and the 1 and 3-screw-(t-butyl PAOKI seesaw propyl) benzene. MFRs were 2.3g / 10 minutes. A presentation and the measurement result of physical properties are shown in Tables 1 and 2.

[0072] In example of comparison 2 example 1, the same actuation as an example 1 was performed except having considered the ethylene amount of supply of this polymerization process 2 as a part for 1.2l./. In addition, the melt flow rates of a 21-mol % and the obtained polymer of the ethylene gas presentation in the gaseous phase at the time of reaction termination were 0.06g / 10min.

[0073] (Accommodation of molecular weight) The same actuation as an example 1 was performed to the obtained polymer except having carried out 0.07 weight section addition of the anti-oxidant 0.2 weight section and the 1 and 3-screw-(t-butyl PAOKI seesaw propyl) benzene. MFRs were 2.9g / 10 minutes. A presentation and a physical-properties measurement result are shown in Tables 1 and 2.

[0074]

[Table 1]

表1

	単量体単位		溶出成分				低結晶成分(A成分＋B成分)の単量体単位		C成分の単量体単位	
	プロピレン mol%	エチレン mol%	A成分 重量%	B成分 重量%	C成分 重量%	D成分 重量%	プロピレン mol%	エチレン mol%	プロピレン mol%	エチレン mol%
実施例1	75	25	37	44	5	14	74	26	42	58
実施例2	79	21	31	50	3	16	76	24	49	51
実施例3	70	30	42	34	8	16	69	31	32	68
比較例1	76	24	36	52.5	0.5	11	74	26	85	15
比較例2	66	34	50	18	12	20	65	35	25	75

[0075]

[Table 2]

表2

	Izod 衝撃値 (−40℃) kJ/m²	曲げ弾性率 MPa	ヘイズ値 %	ビカット 軟化点温度 ℃
実施例1	60	120	18	110
実施例2	51	140	16	112
実施例3	74	100	20	106
比較例1	3.4	120	16	111
比較例2	60以上	89	35	98

[Translation done.]

* NOTICES *

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TECHNICAL FIELD

[Field of the Invention] This invention relates to the outstanding shock resistance and a resin constituent with flexibility, transparency, and thermal resistance good [having especially low-temperature impact resistance].

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PRIOR ART

[Description of the Prior Art] Conventionally, generally the approach of adding ethylene-propylene copolymer rubber, ethylene-butene copolymer rubber, a propylene-butene copolymer, a low consistency, straight chain-like low density polyethylene, etc. as a modifier in crystalline polypropylene is learned as an approach of giving shock resistance to polypropylene.
[0003] However, it could not be satisfied with the conventional constituent which blended such a modifier of shock resistance and transparency at coincidence.
[0004] As compared with that from which the polyolefine system elastomer manufactured by the polymerization method was obtained by the above-mentioned blending method on the other hand, transparency is good. Generally by the manufacture approach by this polymerization method, the two-step polymerization method for performing copolymerization of a propylene and ethylene on a second stage story is performed in the polypropylene component in the first stage story. For example, the method of manufacturing thermoplastic elastomer by the polymerization method is indicated by JP,7-118354,A and JP,10-330430,A, and it is indicated that the propylene ethylene copolymer which has the specific presentation acquired as a result shows good flexibility and transparency.
[0005] However, shock resistance and since especially low-temperature impact resistance was low, as for the polypropylene regin obtained by the above-mentioned approach, the further amelioration was desired.

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EFFECT OF THE INVENTION

[Effect of the Invention] Further, it excels in flexibility, thermal resistance, and transparency, and moreover, fabrication nature is good and the polypropylene resin constituent of this invention can be used suitable for shock resistance especially low-temperature impact resistance, and the various fields for which conventional thermoplastic elastomer is used. For example, in the injection-molding field, each shaping component, such as wheels, such as various packing and a cleaner, a bumper, and a hose, the various shaping components of OA equipment and skiing shoes, a grip, and roller skating are mentioned in the bumper in autoparts, a mud guard, horn putt, lamp packing, and the household-electric-appliances field. On the other hand as a film application, a wrap film, a shrink film, A SUTOTCHIRE film, the film for sealants, a sizing film, Adhesive tape, a masking film, the film for agriculture, a medical-application film pasting base material, As a sheet, the film for adhesive bandages, a surface-protection film, a makeup film, etc. A stationery sheet, An occlusion sheet, a desk mat, the sheet for agriculture, a tarpaulin, the interior epidermis material in autoparts, Corrugated one, a window mall, an air duct hose, a weather toss trip, wire covering material, etc. can be used as a building-materials-related sheet suitable [wallpaper, flooring etc.] for a vanity case, a makeup bag, a shipping box, a package bag, a food container, miscellaneous-goods components, a toy, tatami facing, etc. as a Plastic solid.

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TECHNICAL PROBLEM

[Problem(s) to be Solved by the Invention] Therefore, the purpose of this invention is to have especially low-temperature impact resistance, and for flexibility, transparency, and thermal resistance offer [the outstanding shock resistance and] a good resin constituent.

[Translation done.]

MEANS

[Means for Solving the Problem] In the polypropylene resin constituent which has specific crystal nature cloth, it has especially cold shock-proof nature, and this invention persons came to complete a header and this invention for outstanding being shock resistance and flexibility, transparency, and thermal resistance being good, as a result of repeating research, in order to solve the above-mentioned problem.
[0008] Namely, this invention consists of a polypropylene component and a copolymer component of a propylene and ethylene. (1) A propylene monomeric unit is 90 - 50-mol %, and an ethylene monomeric unit is 10 - 50-mol %. (2) about the leached moiety classified by the temperature temperature up elution separation method using O-dichlorobenzene solvent The amount of 20-degree-C or more less than 85-degree C leached moiety (B component) 20 to 50% of the weight 10 - 70 % of the weight, [the amount of a less than 20-degree C leached moiety (A component)] The amount of 85-degree-C or more less than 100-degree C leached moiety (C component) 3 - 10 % of the weight, The amount of a leached moiety (D component) 100 degrees C or more is 10 - 50 % of the weight. And the sum total of A component, B component, C component, and D component is 100 % of the weight, and a propylene monomeric unit is the polypropylene resin constituent characterized by (3) C components being [a 80 - 20 mol % and ethylene monomeric unit] 20 - 80-mol %.
[0009]
[Embodiment of the Invention] A polypropylene resin constituent consists of a polypropylene component and a copolymer component of a propylene and ethylene in this invention.
[0010] In this invention, a propylene monomeric unit is 90-50-mol %, and an ethylene monomeric unit is 10-50-mol %. If a propylene monomeric unit exceeds 90-mol %, flexibility falls, and in being less than [50 mol %], thermal resistance will fall. When flexibility and heat-resistant balance are taken into consideration, a propylene monomeric unit is desirable and a 88-52-mol % and ethylene monomeric unit is 12-48-mol %. Setting to this invention, temperature temperature up elution separation method (TREF) is Journalof. Applied Polymer Science;AppliedPolymer Symposium It is based on 45 and the approach described by the detail 1-24 (1990).
[0011] That is, it is the approach of carrying out elution to order from a component with the low melting point, and isolating an elution polymer component preparatively, by introducing a hot polymer solution into the column which used diatomaceous earth as a bulking agent first, crystallizing a bulking agent front face sequentially from a component with the high melting point, and then raising column temperature gradually by reducing column temperature gradually. Therefore, in this specification, less than 20-degree C leached moieties are all leached moieties in the temperature up process in which it results [from a low temperature side] in 20 degrees C, in temperature temperature up elution separation method.
[0012] In this invention, the amount of the less than 20-degree C leached moiety (it abbreviates to A component hereafter) classified by the above-mentioned temperature temperature up elution separation method is 20 - 50 % of the weight. If shock resistance and flexibility are spoiled and A component exceeds 50 % of the weight at 20 or less % of the weight, thermal resistance will fall. When shock resistance, flexibility, and heat-resistant balance are taken into consideration, the amount of A components is 25 - 45 % of the weight preferably.

[0013] In this invention, the amount of 20-degree-C or more less than 85-degree C leached moiety (it abbreviates to B component hereafter) is 10 - 70 % of the weight. When the amount of B components is less than 10 % of the weight, shock resistance, flexibility, and transparency fall. Moreover, thermal resistance cannot be satisfied if it exceeds 70 % of the weight. In order to have the outstanding shock resistance and flexibility, transparency, and thermal resistance, the amount of B components is 15 - 65 % of the weight preferably.
[0014] In this invention, a propylene monomeric unit is [the 90-50 mol % and ethylene monomeric unit of the low crystal component which consists of an A component and a B component] 10-50-mol %. When flexibility falls when an ethylene monomeric unit is less than [10 mol %], and an ethylene monomeric unit exceeds 50-mol %, thermal resistance falls. if flexibility and heat-resistant balance are taken into consideration -- the ethylene monomeric unit of a low crystal component -- desirable -- 12-48-mol % -- it is 15-45-mol % more preferably. In this invention, the amount of 85-degree-C or more less than 100-degree C leached moiety (it abbreviates to C component hereafter) is 3 - 10 % of the weight. When the amount of C components is less than 3 % of the weight, shock resistance, especially low-temperature impact resistance fall. Moreover, when exceeding 10 % of the weight, transparency falls. When shock resistance and transparency are taken into consideration, the amount of C components is 3 - 9 % of the weight preferably. In this invention, a propylene monomeric unit is [the 80-20 mol % and ethylene monomeric unit of C component] 20-80-mol %. When the propylene monomeric unit of C component is less than [20 mol %], thermal resistance falls, and in exceeding 80-mol %, shock resistance, especially low-temperature impact resistance fall. if thermal resistance and shock-proof balance are taken into consideration -- the ethylene monomeric unit of C component -- desirable -- 20-75-mol % -- it is 25-70-mol % more preferably. The propylene-ethylene random-copolymer component and ethylene monomeric unit beyond 75 mol % are considered that C component consists of the so-called block copolymer which the propylene-ethylene copolymer of the two above-mentioned component arranged [the propylene monomeric unit] in mixture with the propylene-ethylene random-copolymer component beyond 90 mol %, or a monad chain here. In this invention, it is thought [shock resistance and] that an ethylene monomeric unit depends especially the improvement in low-temperature impact resistance on the propylene-ethylene random-copolymer component beyond 90 mol %.
[0015] In this invention, the amount of a leached moiety (henceforth D component) 100 degrees C or more is 10 - 50 % of the weight. When the amount of D components is less than 10 % of the weight, while the increase of the adhesiveness of the polypropylene resin powder obtained and manufacture become difficult, it is not desirable in order for thermal resistance to fall. Moreover, when exceeding 50 % of the weight, since shock resistance and flexibility fall, it is not desirable. When productivity, thermal resistance, shock resistance, and flexibility are taken into consideration, the amount of D components is less than 30 % of the weight 10 % of the weight or more more preferably ten to 45% of the weight.
[0016] In this invention, a propylene monomeric unit is the fraction which almost accounts for a total rate, and D component consists of a propylene-ethylene random copolymer to which a 100-95-mol % and ethylene monomeric unit changes [a propylene monomeric unit] from 0-5-mol %.
[0017] In this invention, the sum total of the above-mentioned A component, B component, C component, and D component is 100 % of the weight.
[0018] The manufacture approach of the polypropylene resin constituent of this invention is not limited especially as long as the requirements for this invention are satisfied.
[0019] For example, the approach of manufacturing by carrying out the polymerization of the component of the mode which is independent, respectively about the well-known resin which uses the component of - (D) as a principal component, respectively, or is manufactured by blending so that the conditions of this invention may be satisfied for the well-known resin which contains two or more sorts of the above-mentioned component at a rate of arbitration or (A) (A) - (D) which has said property continuously or gradually is mentioned.
[0020] Among those, the approach of acquiring by carrying out a polymerization continuously or

gradually improves the dispersibility of each component, and it is the most desirable in order to discover especially cold shock nature enough, the outstanding shock resistance and. Specifically, the method of obtaining the so-called block copolymer which carried out the polymerization of a propylene-ethylene random-copolymer component while the polypropylene component changed the propylene / ethylene presentation continuously or gradually after the polymerization, and the polypropylene component and the copolymerization component of a propylene and ethylene arranged in distribution or a monad chain micro is desirable in order to demonstrate shock-proof improvement, in addition good transparency.

[0021] The following approaches will be mentioned if the typical approach of the approach of manufacturing the polypropylene resin constituent of this invention by the multistage polymerization is illustrated.

[0022] That is, after carrying out the polymerization of the propylene to the bottom of existence of the following catalyst component [I], [RO], and [Ha], it is the approach of performing random copolymerization of a propylene and ethylene succeedingly.

[**] A titanium compound with well-known the titanium compound [RO] organoaluminium compound [Ha] organic silicon compound above-mentioned titanium compound [I] being used for the polymerization of an olefin is used that there is no limit in any way. Especially, when it is used for the polymerization of a propylene, the titanium compound which can obtain the polymer of high stereoregularity by high yield is desirable. These titanium compounds are divided roughly into a support mold titanium compound and a titanium-trichloride compound. The process of a support mold titanium compound is adopted that a well-known approach does not have a limit in any way. For example, JP,56-155206,A, said -- 56-136806 -- said -- 57-34103 -- said -- 58-8706 -- said -- 58-83006 -- said -- 58-138708 -- said -- 58-183709 -- said -- 59-206408 -- said -- 59-219311 -- said -- 60-81208 -- said -- 60-81209 -- said -- 60-186508 -- said -- 60-192708 -- said -- 61-211309 -- said -- 61-271304 -- said -- 62-15209 -- said -- 62-11706 -- said -- the approach shown in 62-72702 and this 62-104810 grade is adopted. Specifically, the approach of co-grinding halogenation titanium and a magnesium compound or the method of contacting halogenation titanium, a magnesium compound, and an electron donor in a solvent is listed to the bottom of existence of electron donors, such as the approach of co-grinding a titanium tetrachloride with a magnesium compound like a magnesium chloride, alcohol, the ether, ester, a ketone, or an aldehyde.

[0023] Moreover, alpha, beta, gamma, or delta-3 titanium chloride well-known as a titanium-trichloride compound is mentioned. the preparation approach of these titanium-trichloride compounds -- for example, JP,47-34478,A -- said -- 50-126590 -- said -- 50-114394 -- said -- 50-93888 -- said -- 50-123091 -- said -- 50-74594 -- said -- 50-104191 -- said -- 50-98489 -- said -- 51-136625 -- said -- the approach shown in 52-30888 and this 52-35283 grade is adopted. In order especially to obtain the propylene-ethylene copolymer of this invention also in this, a titanium-trichloride compound is desirable.

[0024] Next, a compound with well-known an organoaluminium compound [RO] being used for the polymerization of an olefin is adopted that there is no limit in any way. For example, trimethylaluminum, triethylaluminum, tree n propyl aluminum, Tree n butyl aluminum, tree i butyl aluminum, tree n hexyl aluminum, Trialkylaluminiums, such as tree n octyl aluminum and tree n DESHIRU aluminum; Diethyl aluminum mono-chloride, Diethyl aluminum mono-halide, such as a diethyl aluminum star's picture; alkylaluminum halide, such as methyl aluminiumsesquichloride, ethylaluminiumsesquichloride, and diethyl aluminum chloride, is mentioned. Alkoxy aluminum, such as mono-ethoxy diethyl aluminum and diethoxy monoethyl aluminum, can be used for others. Also in this, trialkylaluminiums especially trimethylaluminum, triethylaluminum, and tree i butyl aluminum are desirable.

[0025] furthermore, **, such as a cyclic hydrocarbon which an organic silicon compound [Ha] is a chain-like hydrocarbon whose atom linking directly to a silicon atom is the 3rd class carbon although a compound with well-known being used for stereoregularity amelioration of an olefin is adopted that there is no limit in any way, or is the 2nd class carbon, -- in order that the organic silicon compound which has a high substituent may make the stereoregularity of the polypropylene component obtained higher and may discover good thermal resistance, it is

desirable. Specifically JI t-butyl dimethoxysilane, t-butyl ethyl dimethoxysilane, JI t-amyl dimethoxysilane, dicyclopenthyl dimetoxysilane, Dicyclohexyl dimetoxysilane, t-butyl methyl dimethoxysilane, t-butyl ethyl dimethoxysilane, cyclopentyl methyl dimethoxysilane, Organic silicon compounds, such as cyclopentyl ethyl dimethoxysilane, cyclopentyl isobutyl dimethoxysilane, cyclohexyl methyldimetoxysilane, cyclohexyl ethyl dimethoxysilane, and cyclohexyl isobutyl dimethoxysilane, can be mentioned. Dicyclopenthyl dimetoxysilane and especially t-butyl ethyl dimethoxysilane are desirable especially. Moreover, these organic silicon compounds can also use two or more sorts for coincidence.

[0026] the combination of the titanium compound [I] used for this invention, an organoaluminium compound [RO], and an organic silicon compound [Ha] -- (1) titanium-trichloride compound-trialkylaluminium -- a kind-organic silicon compound (2) support mold titanium compound-trialkylaluminium-organic silicon compound (3) -- the combination of a **** type titanium compound-3 titanium-chloride compound-trialkylaluminium-organic silicon compound is desirable especially in order to satisfy the requirements for a configuration of the polypropylene resin constituent of this invention. Especially the combination of (1) is suitable in order to obtain the polypropylene resin constituent with which are satisfied of this invention.

[0027] Since the smeariness at the time of setting to this invention, and performing precuring by mixing with propylene independence, ethylene independence or a propylene, and ethylene to the bottom of existence of the above-mentioned catalyst component [I], [RO], and [Ha] reducing the amount of generation of the low-molecular-weight object of the polypropylene resin constituent obtained in advance of this polymerization under existence of each above-mentioned component, and considering as mold goods can be suppressed, it is suitable. The iodine compound which is furthermore shown by the following general formula if needed in addition to the above [I], [RO], and [Ha] [NI]

Since performing precuring by mixing with propylene independence, ethylene independence or a propylene, and ethylene to the bottom of existence of R-I (however, R being the alkyl group or phenyl group of an iodine atom or carbon numbers 1-7.) can suppress further the smeariness at the time of controlling further generation of the above-mentioned low-molecular-weight object, and using the polypropylene resin constituent obtained as a Plastic solid, it serves as a desirable mode.

[0028] Since the amounts of the above [I] used by precuring of this invention, [RO] and [Ha], and each catalyst component of [NI] used further if needed differ according to the class of catalyst component, and the conditions of a polymerization, they should just determine the optimal amount used beforehand according to these monograph affairs. It will be as follows if the range used suitably is illustrated.

[0029] the operating rate of the organoaluminium compound [RO] used for precuring -- a titanium compound [I] -- receiving -- aluminum/Ti (mole ratio) -- 0.1-100 -- desirable -- the range of 0.1-20 -- the operating rate of an organic silicon compound [Ha] -- a titanium compound [I] -- receiving -- [Ha]/Ti (mole ratio) -- 0.01-100 -- the range of 0.01-10 is preferably suitable respectively. moreover, the operating rate of the iodine compound [NI] used if needed -- a titanium compound [I] -- receiving -- I/Ti (mole ratio) -- 0.1-100 -- the range of 0.5-50 is preferably suitable.

[0030] It is as follows when the iodine compound which can be suitably used by precuring of this invention is shown concretely. For example, they are iodine, a methyl iodide, an ethyl iodide, propyl iodide, iodation butyl, an iodobenzene, p-iodation toluene, etc. Especially a methyl iodide and an ethyl iodide are suitable.

[0031] although the amount of precuring which carries out a polymerization by mixing of propylene independence, ethylene independence or a propylene, and ethylene to the bottom of existence of said catalyst component changes with precuring conditions -- general -- a 0.1 - 250 g/g-Ti compound -- if it is the range of a 1 - 100 g/g-Ti compound preferably, it will come out enough.

[0032] It is possible in the case of precuring to also make hydrogen live together. As for this precuring, it is desirable to make a slurry polymerization usually apply, and as a solvent, it is independent about saturated aliphatic hydrocarbon or aromatic hydrocarbon, such as a hexane, a

heptane, a cyclohexane, benzene, and toluene, or can use these mixed solvents.
[0033] This especially precuring temperature has the desirable range of 0-60 degrees C -20-100 degrees C. What is necessary is just to decide on precuring time amount suitably according to the lap in precuring temperature and precuring. Although the pressure in precuring is not limited, in the case of a slurry polymerization, generally, it is about atmospheric pressure -5 kg/cm2G. This precuring may be performed by the batch, the half-batch, and which approach of continuation.
[0034] Subsequently to said precuring, this polymerization is carried out. Under existence of the catalyst content precuring object with which this polymerization was obtained by said precuring, the polymerization of a polypropylene component is performed first and then random copolymerization of propylene-ethylene is carried out. Moreover, although each catalyst component can also use what was added at the time of precuring in the condition as it is, it is desirable except a titanium compound to newly add and to adjust at the time of this polymerization.
[0035] Since the amount and polymerization conditions of each catalyst component of the above [I] used by this polymerization of this invention, [RO], and [Ha] differ from each other according to the class of catalyst component, they should just determine the optimal amount used and polymerization conditions beforehand according to the class of these catalyst components. It will be as follows if the amount and polymerization conditions of a catalyst component which are used suitably are illustrated.
[0036] The above-mentioned thing can use the organoaluminium compound [RO] used by this polymerization that there is no limit in any way. the amount of the organoaluminium compound used -- catalyst content precuring -- a titanium atom in the living body -- receiving -- aluminum/Ti (mole ratio) -- it is -- 1-1000 -- it is 2-500 preferably. Moreover, although this lap changes with these polymerization conditions, generally it is the range of 1000 - 50000 g/g and a titanium compound.
[0037] The organic silicon compound [Ha] used by this polymerization is adopted that a compound as stated above does not have a limit in any way. the amount of the organic silicon compound used used by this polymerization -- catalyst content precuring -- a titanium atom in the living body -- receiving -- Si/Ti (mole ratio) -- 0.001-1000 -- it is 0.1-500 preferably.
[0038] As for this above-mentioned polymerization, the polymerization of a polypropylene component is carried out first. The polymerization of a polypropylene component supplies the propylene in within the limits and the mixture of ethylene with which are satisfied of the requirements for a propylene independent or this invention, and should just carry them out. If the typical conditions of a polypropylene component polymerization are illustrated, it is suitable for polymerization temperature to adopt from the range of 80 degrees C or less and further 20-70 degrees C. Moreover, hydrogen can also be made to live together as a molecular weight modifier if needed. Furthermore, which approaches, such as a slurry polymerization which uses the propylene itself as a solvent, a vapor phase polymerization, and solution polymerization, may be used for a polymerization again. When the shape of the simple nature of a process, a reaction rate, and corpuscular character of a copolymer to generate is taken into consideration, the slurry polymerization which uses the propylene itself as a solvent is a desirable mode. Which approach of a batch process, a half-batch process, and continuous system is sufficient as a polymerization format. Furthermore, a polymerization can be divided into two or more steps where conditions, such as hydrogen concentration and polymerization temperature, differ, and can also be performed.
[0039] Next, random copolymerization of a propylene and ethylene is performed. Which approach of a batch process, a half-batch process, and continuous system may be used for the random copolymerization of a propylene and ethylene, and it can divide a polymerization into a multistage story and can also carry it out. Moreover, which approach of a slurry polymerization, a vapor phase polymerization, and solution polymerization may be used for the polymerization of this process. It is continuing at the polymerization of said polypropylene component in the case of the slurry polymerization which uses the propylene itself as a solvent, and supplying ethylene gas, and, in the case of a vapor phase polymerization, carries out by supplying the mixed gas of a

propylene and ethylene.
[0040] In order to satisfy the weight rate of (A) - (D) component of this invention especially, it is desirable to perform random copolymerization of a propylene and ethylene by the slurry polymerization following the polymerization of a polypropylene component. In that case, ethylene's supplying ethylene improves the dispersibility of each component so that ethylene / propylene presentation ratio may go up continuously, and supply, or ethylene / propylene presentation ratio may change a constant rate gradually continuously, and it is desirable in order to fully demonstrate especially the improvement in a cold shock, in addition good transparency, shock resistance and.
[0041] 80 degrees C or less of polymerization temperature of the random copolymerization of a propylene and ethylene are preferably adopted from the range of 20-70 degrees C. Moreover, hydrogen can also be used as a molecular weight modifier if needed, and the hydrogen concentration in that case can be changed to a multistage story, and can also carry out a polymerization.
[0042] After termination of this polymerization, a monomer can be separated from a polymerization system and the polypropylene resin constituent of this invention can be obtained. This polypropylene resin constituent can perform well-known washing or counterflow washing with a with a carbon number of seven or less hydrocarbon.
[0043] In this invention, although especially the melt flow rate of a polypropylene resin constituent is not limited, generally it is desirable that they are 0.1-100g / 10min. When the above-mentioned melt flow rates are 0.1g / less than 10 min, a moldability falls, when exceeding 100g / 10min, a low-molecular-weight object increases and the fall of physical properties may be caused. if a moldability and physical properties are taken into consideration -- a melt flow rate -- desirable -- 0.1-70g/-- they are 0.1-50g / 10min still more preferably 10 min.
[0044] The melt flow rate can be adjusted and the polypropylene resin constituent of this invention can also be used so that it may become the melt flow rate demanded by the application etc. if needed. The method of adding organic peroxide to the polypropylene resin powder which might make a small amount of hydrogen live together as an approach of adjusting a melt flow rate, at the time of a polymerization, and adjusting molecular weight by carrying out melting kneading is desirable in respect of reduction of a low-molecular-weight object, and the improvement in a moldability.
[0045] After adding fillers, such as commercial additives, such as an antioxidant, a coloring agent, a thermostabilizer, a chlorine supplement agent, lubricant, an ultraviolet ray absorbent, an illuminant-proof, an antistatic agent, an antifogger, an anti blocking agent, a flame retarder, and a crystalline-nucleus agent, and talc, a magnesium hydroxide, a calcium carbonate, glass, a mica, wood flour, and mixing, you may use by making the polypropylene resin constituent of this invention into a pellet with an extruder.
[0046] Furthermore, to the polypropylene resin constituent of this invention, other resin can be added in the range which does not affect the property of this invention remarkably. For example, alpha olefins more than 90% mol of a propylene, and other than a propylene, For example, 1-butene, 1-pentene, 1-hexene, 1-heptene, The random copolymer not more than [one or more sorts of /, such as 1-methyl-1-pentene,] 10 mol %, High density polyethylene (HDPE), low density polyethylene (LDPE), the line which becomes by copolymerization of ethylene, and C4-C10 -- polyethylene (LLDPE) -- Polyethylene system resin, such as ethylene vinyl acetate copolymerization (EVA) and ethylene methacrylate (EMMA), Ethylene propylene rubber (EPR, EPDM), an ethylene butene-1 copolymer (EBM), Well-known things, such as olefin system elasticity resin, a styrene butadiene block copolymer (SBR), petroleum resin and terpene resin, or those hydrogenation objects, such as a propylene butene-1 copolymer (PBM), can use it without a limit.
[0047] As mentioned above, of course, it can also be used as a modifier of various polyolefines besides other polypropylene regins using properties, such as the property, for example, flexibility, and transparency, that the polypropylene resin constituent of this invention can make this a principal component, and it can be used for a shaping raw material.
[0048] The resin constituent of the polypropylene resin constituent of this invention and other

resin is obtained by carrying out melting kneading. Although especially the approach of melting kneading is not limited, it is good to, perform 160-300 degrees C to the bottom of the temperature of 180-270 degrees C preferably for example, using a screw extruder, a Banbury mixer, a roll mill, etc. Moreover, this melting kneading can also be performed under inert gas air currents, such as nitrogen gas. In addition, well-known mixed equipment, for example, a tumbler, a Henschel mixer, etc. can use it before melting kneading that there is no limit in any way.
[0049] Furthermore, it is also possible by mixing it, after each component of the above-mentioned resin constituent etc. blends each component if needed, supplying to a direct making machine and fabricating to acquire a Plastic solid. Moreover, when fabricating a film sheet etc., it can fabricate by the well-known shaping approach. For example, the shaping approaches, such as T-die shaping, calender shaping, and inflation molding, can be used.
[0050] It can also be used by the monolayer besides using it as a film sheet, the polypropylene resin constituent of this invention being able to carry out the laminating of the resin layers, such as ethylene system resin, such as propylene system resin, high density polyethylene, low density polyethylene, straight chain-like low density polyethylene, an ethylene-vinylacetate copolymer, and an ethylene-methacrylic-acid copolymer, and multilayering it.

[Translation done.]

* NOTICES *

EXAMPLE

[Example] Hereafter, although an example and the example of a comparison are given and this invention is explained, this invention is not limited to these examples. The measuring method used in the following examples is explained.
[0053] 1) the measurement JEOL [Co., Ltd.] Co., Ltd. make of a propylene monomeric unit and an ethylene monomeric unit -- it computed from 13 C-NMR spectrum measured using GSX-270.

[0054] 2) The judgment approach of A component, B component, C component, and D component (the temperature temperature up elution judgment approach)
The following Measuring conditions performed using MPC[by the YUNIFU Rose company]-1100 mold.
[0055]
Solvent 30 - 60mesh column cooling rate; 5 degrees C/Hr The;O-dichlorobenzene rate of flow ; 1.0 ml/min programming rate ; 40 degrees C / Hr sample concentration ; 0.5wt% sample injection rate; 500microl detector ; An infrared detector, wavelength column of 3.14 micrometers ; phi0.9cmx17.5cm bulking agent ; chromosrb [0056] P 3) Measurement ASTM of a melt flow rate (MFR) It was based on D-1238.
[0057] 4) Shock-proof (Izod impact resistance value) measurement JIS Based on K7110, it measured with the notch in the temperature of -40 degrees C.
[0058] 5) Measurement JIS of flexibility (bending elastic modulus) It was based on K7203.
[0059] 6) Measurement JIS of transparency (Hayes value) It was based on K6714.
[0060] 7) Heat-resistant (BIGATTO softening temperature) measurement JIS Based on K7206, it measured on the conditions of 250g of loads.
[0061] The example of the polypropylene resin constituent of this invention is explained below.
[0062] After nitrogen gas fully permuted the glass autoclave reactor of 1l. of content volume equipped with example 1 (precuring) agitator, hexane 400ml was inserted in. After keeping whenever [reactor internal temperature] at 20 degrees C and adding dicyclopenthyl dimetoxysilane 4.2mmol, ethyl-iodide 21.5mmol, triethylaluminum 21.5mmol, and titanium-trichloride (Marubeni Solvay chemistry company make) 21.5mmol, the propylene was continuously introduced into the reactor for 30 minutes so that it might be set to 3g per 1g of titanium trichlorides. In addition, temperature in the meantime was held at 20 degrees C. After suspending supply of a propylene, nitrogen gas fully permuted the inside of a reactor, and the obtained titanium content polypropylene was washed 4 times by the purification hexane. The polymerization of the 2.7g [per 1g of titanium trichlorides] propylene was carried out as a result of analysis.
[0063] (This polymerization) In addition, the temperature up of the internal temperature of an autoclave was carried out to 55 degrees C so that the concentration in a gaseous phase might become the 2l. autoclave which performed N2 permutation about a liquid propylene and might become 1.0-mol% about 1l., triethylaluminum 0.5mmol, dicyclopenthyl dimetoxysilane 0.25mmol, and hydrogen. Next, after supplying ethylene so that the concentration in a gaseous phase may become 1.5-mol%, in addition, copolymerization of propylene-ethylene was performed for 20 minutes at 55 degrees C 0.06 mmols by making into a titanium trichloride the titanium content

polypropylene obtained by precuring (process 1). Subsequently, ethylene was supplied by part for 0.8l./by reference condition, and the polymerization for 120 minutes was performed (process 2). The rate of ethylene gas to the propylene gas in a gaseous phase increased as propylene-ethylene copolymerization time amount made it the increment. In addition, the ethylene gas presentation in the gaseous phase at the time of reaction termination was 15-mol%. After reaction termination, the unreacted monomer was purged and the polymer was obtained. The obtained polymer was dried at 70 degrees C for 1 hour. The melt flow rates of the obtained polymer were 0.10g / 10min.

[0064] (Molecular weight accommodation) As the 0.2 weight section and organic peroxide, the anti-oxidant was extruded at 230 degrees C using Brabender to it, after carrying out 0.05 weight section addition of the 1 and 3-screw-(t-butyl PAOKI seesaw propyl) benzene and mixing to the obtained polymer, and the pellet was obtained. MFRs were 2.0g / 10 minutes.

[0065] (Component analysis) Measurement of the propylene monomeric unit of the low crystal component which consists of measurement of the propylene monomeric unit of a polypropylene resin constituent and an ethylene monomeric unit, A-D each amount measurement of components, an A component, and a B component, and D component, and an ethylene monomeric unit was performed using the pellet which may have had molecular weight accommodation performed. A result is shown in Table 1.

[0066] (Physical-properties measurement) The test piece for Hayes with a thickness of 1mm was fabricated with 15t injection molding machine using the pellet which may have had molecular-weight accommodation performed with the cylinder temperature of 230 degrees C, and the die temperature of 40 degrees C in a bending modulus of elasticity, BIGATTO softening temperature and the shock-proof test piece, and the list. The measurement result of physical properties is shown in Table 2.

[0067] In molecular-weight adjustment of example 2 example 1, the same actuation as an example 1 was performed except having considered the ethylene amount of supply of this polymerization process 2 as a part for 0.6l./. In addition, the melt flow rates of a 12-mol % and the obtained polymer of the ethylene gas presentation in the gaseous phase at the time of reaction termination were 0.12g / 10min. A result is shown in Tables 1 and 2.

[0068] In example 3 example 1, the same actuation as an example 1 was performed except having considered the ethylene amount of supply of this polymerization process 2 as a part for 1.0l./. In addition, the melt flow rates of a 18-mol % and the obtained polymer of the ethylene gas presentation in the gaseous phase at the time of reaction termination were 0.08g / 10min. A result is shown in Tables 1 and 2.

[0069] After nitrogen gas fully permuted the glass autoclave reactor of 1l. of content volume equipped with example of comparison 1 (precuring) agitator, hexane 400ml was inserted in. After keeping whenever [reactor internal temperature] at 20 degrees C and adding diethyl aluminum chloride 18.5mmol, ethyl-iodide 22.7mmol, diethylene-glycol wood ether 0.18mmol, and titanium-trichloride (Marubeni Solvay chemistry company make) 22.7mmol, the propylene was continuously introduced into the reactor for 30 minutes so that it might be set to 3g per 1g of titanium trichlorides. In addition, temperature in the meantime was held at 20 degrees C. After suspending supply of a propylene, nitrogen gas fully permuted the inside of a reactor, and the obtained titanium content polypropylene was washed 4 times by the purification hexane. The polymerization of the 2.9g [per 1g of titanium trichlorides] propylene was carried out as a result of analysis.

[0070] (This polymerization) In addition, the temperature up of the internal temperature of an autoclave was carried out to 55 degrees C so that the concentration in a gaseous phase might become the 2l. autoclave which performed N2 permutation about a liquid propylene and might become 1.0-mol% about 1l., diethyl aluminum chloride 0.7mmol, butyl-acetate 0.07mmol, dicyclopenthyl dimetoxysilane 0.07mmol, and hydrogen. Next, after supplying ethylene so that the concentration in a gaseous phase may become 1.0-mol%, in addition, copolymerization of propylene-ethylene was performed for 30 minutes at 55 degrees C 0.087 mmols by making into a titanium trichloride the titanium content polypropylene obtained by precuring (process 1). Subsequently, ethylene was supplied by part for 0.5l./by reference condition, and the

polymerization for 120 minutes was performed (process 2). The rate of ethylene gas to the propylene gas in a gaseous phase increased as propylene-ethylene copolymerization time amount made it the increment. In addition, the ethylene gas presentation in the gaseous phase at the time of reaction termination was ten-mol%. After reaction termination, the unreacted monomer was purged and the polymer was obtained. The obtained polymer was dried at 70 degrees C for 1 hour. The melt flow rates of the obtained polymer were 0.03g / 10min.

[0071] (Molecular-weight accommodation) The same actuation as an example 1 was performed to the obtained polymer except having carried out 0.08 weight section addition of the anti-oxidant 0.2 weight section and the 1 and 3-screw-(t-butyl PAOKI seesaw propyl) benzene. MFRs were 2.3g / 10 minutes. A presentation and the measurement result of physical properties are shown in Tables 1 and 2.

[0072] In example of comparison 2 example 1, the same actuation as an example 1 was performed except having considered the ethylene amount of supply of this polymerization process 2 as a part for 1.2l./. In addition, the melt flow rates of a 21-mol % and the obtained polymer of the ethylene gas presentation in the gaseous phase at the time of reaction termination were 0.06g / 10min.

[0073] (Accommodation of molecular weight) The same actuation as an example 1 was performed to the obtained polymer except having carried out 0.07 weight section addition of the anti-oxidant 0.2 weight section and the 1 and 3-screw-(t-butyl PAOKI seesaw propyl) benzene. MFRs were 2.9g / 10 minutes. A presentation and a physical-properties measurement result are shown in Tables 1 and 2.

[0074]

[Table 1]

表1

	単量体単位		溶出成分				低結晶成分(A成分＋B成分)の単量体単位		C成分の単量体単位	
	プロピレン mol%	エチレン mol%	A成分 重量%	B成分 重量%	C成分 重量%	D成分 重量%	プロピレン mol%	エチレン mol%	プロピレン mol%	エチレン mol%
実施例1	75	25	37	44	5	14	74	26	42	58
実施例2	79	21	31	50	3	16	76	24	49	51
実施例3	70	30	42	34	8	16	69	31	32	68
比較例1	76	24	36	52.5	0.5	11	74	26	85	15
比較例2	66	34	50	18	12	20	65	35	25	75

[0075]

[Table 2]

表2

	Izod 衝撃値 (−40℃) kJ/m²	曲げ弾性率 MPa	ヘイズ値 %	ビカット 軟化点温度 ℃
実施例1	60	120	18	110
実施例2	51	140	16	112
実施例3	74	100	20	106
比較例1	3.4	120	16	111
比較例2	80以上	89	35	98

[Translation done.]

* NOTICES *

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DESCRIPTION OF DRAWINGS

[Brief Description of the Drawings]
[Drawing 1] Drawing 1 is the elution diagram of the temperature temperature up elution separation method of the propylene ethylene block-copolymer constituent of an example 1.
[Drawing 2] Drawing 2 is the elution diagram of the temperature temperature up elution separation method of the propylene ethylene block-copolymer constituent of the example 1 of a comparison.

[Translation done.]

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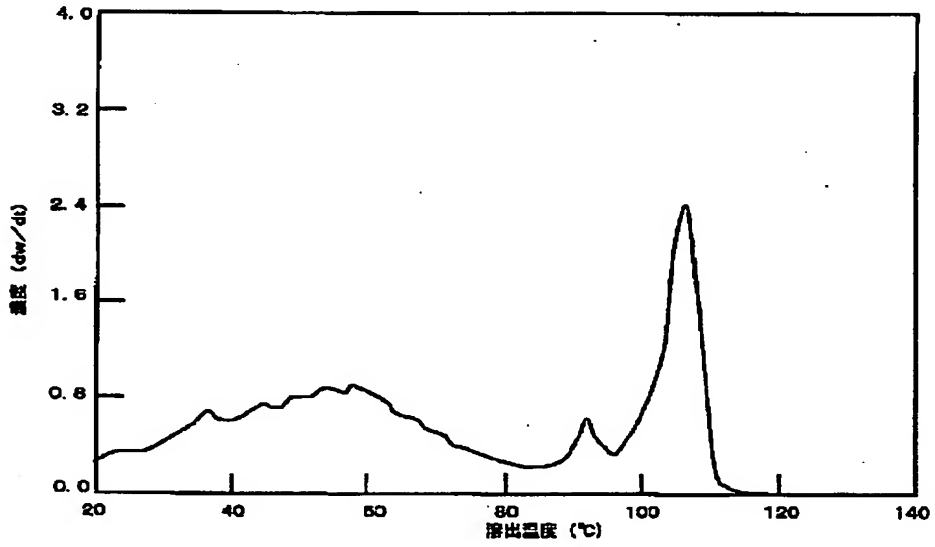
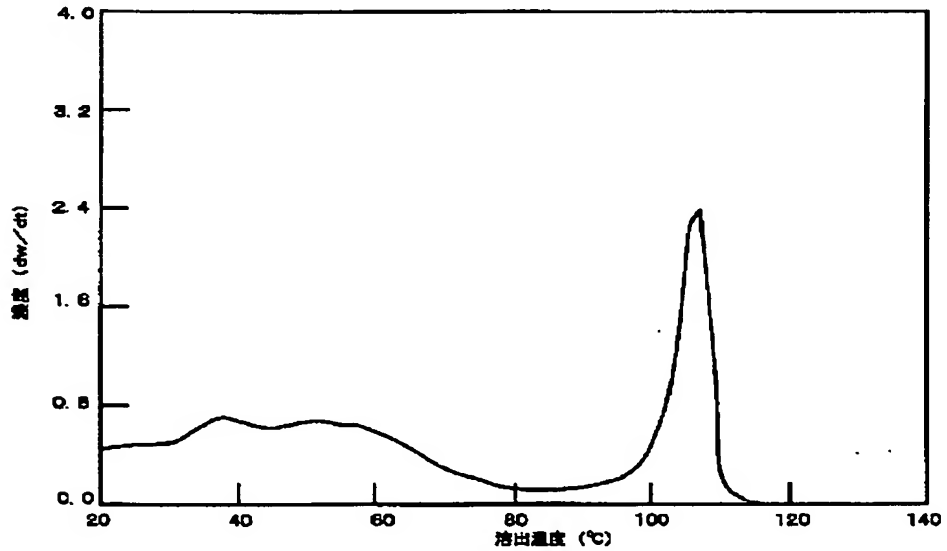
DRAWINGS

[Drawing 1]
図1



[Drawing 2]
図2



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